FIG. 1: Phase diagram of $\lambda-(BETS)_2FeCl_4$. The four phases identified are antiferromagnetic (AF), canted antiferromagnetic (CAF), paramagnetic metal (PM), and field induced superconducting (FISC) states. Closed (open) symbols represent the temperature (field) sweep measurements of the CAF sub-phase structure: A-circles, B-squares, C-triangles (see text). Schematic: Top figure: sample geometry and principle axes. Lower figures: axial and side views of sample orientation. The sample position shown is for the c-axis$\parallel\hat{r}$ and the c-axis$\perp\hat{z}$, where $\hat{r}$ and $\hat{z}$ are the unit directions for the axial and radial components of the cylindrical cavity.

tions, is that the CAF spin structure appears to change in steps as the field (and/or temperature) is increased, and that these discrete changes in spin structure cause corresponding steps in the complex conductivity as the PM phase is approached.

Since the resistance below T_N rapidly becomes unmeasurable, we have probed the antiferromagnetic phase through the use of ac magnetoconductivty, and electron magnetic resonance in the 40 to 110 GHz frequency range. In previous X-band studies[5], the temperature dependent electron spin (ESR) and antiferromagnetic (AFR) resonance features (below T_N) have been investigated vs. temperature and sample orientation with respect to magnetic field. In the present study we extend the range of investigation of the CAF state (and also the

FIG. 2: Low temperature ($T < T_N$) field-dependent mm wave cavity response of $\lambda-(BETS)_2FeCl_4$ for $c \parallel \hat{z}$, $c \parallel \vec{H}$. (All data is for $\nu = 66.9$ GHz, except the 0.5 K data in the main panel where $\nu = 75.4$ GHz.) The sharp dip near 2 T is the ESR line, and the features in the range 5 to 10 T arise from sub-phases in the antiferromagnetic CAF state (see text). Although the sample was aligned to allow the high field superconducting (FISC) state to be stabilized, only weak features associated with a metal to superconducting transition were detected. Inset: Full field sweeps to 11 T. Hysteresis is observed in the sub-phase structure in the CAF state.

FISC state) to a broader range of frequencies, temperatures, and magnetic fields. A mm wave resonant cavity perturbation method is employed[7] where the sample orientation in the cavity can be changed with respect to the field orientation. Samples were synthesized by electrochemical methods[8]. The single crystal used in this work, which was 0.97x0.10x0.10 mm^3 in size, was greased to a small teflon mount inside the cylindrical cavity resonator. In Fig. 1 the convention for orientation is given in terms of the principle axes of the crystal, and the radial and axial ($\hat{z} \parallel \vec{H}$ in all cases) unit vectors associated with the cavity. A control experiment insured that the grease and teflon support did not contribute a significant background or spurious ESR signal.

Experiments were carried out in a 30 T resistive magnet and an 8 T superconducting magnet where either temperature or field were held fixed at a specific resonant cavity mode(frequency). A helium 3-probe was modified to allow temperatures down to 0.5 K without interference from liquid dielectric effects. Our results are presented in Figs. 2-5, and the phase diagram of the CAF state, based on all of our results, is shown in Fig. 1. Representative results are shown for magnetic field sweeps in Fig. 2 for $c \| \hat{z}$ for several temperatures below T_N up to 30 T. The signal is the phase-locked amplitude of the cavity response, where resonant frequency is allowed to change (via feedback) to keep the phase reference in quadrature. In general, there are two signals in the mm wave cavity response, a <u>resonant signal</u> (i.e. $h\nu = g\mu B$) due to electron spin, and <u>non-resonant structure</u> where there is no functional relationship between frequency and field position. The latter arises from changes in the ac complex conductivity of the material. For increasing field an ESR adsorption line appears, followed by a change in background signal that is punctuated by dips, peaks, and/or shoulders. The field positions of the CAF-PM and PM-FISC phase boundaries observed in transport data are also shown in Fig. 2 for comparison. The features at higher field are not resonant, but reflect step-like changes in the ac magnetoconductivity. As discussed below, we interpret these features as sub-phase transitions in the CAF state for increasing field. An exploration of the high field region where the field induced superconducting phase is stabilized did not show any systematic signature, other than slight, hysteretic loop-like features that were not readily reproducible.

We first address the magnetic resonance data, that is the ESR and AFR measurements used to determine the g-factor and spin-flop field respectively. A temperature dependent investigation of the ESR line, shown in Fig. 3, reveals similar features to those previously reported by Brossard *et al.*, with the g-value exhibiting a non-monotonic temperature dependence below T_N. The linear frequency-field dependence of the ESR line yielded a g-value of 2.05 for sample orientations away from those where the AFR signal was observable, as shown in Fig. 3b.

For the orientation $a \| \vec{H}, c \| \hat{r}$, we measured the field position of the the AFR signal over our accessible range of frequency, as plotted in Fig. 3b (see also representative trace of data for $\| \vec{H}, c \| \hat{r}$ data in Fig. 5 below). Although the frequency dependence of the AFR signal is characteristic of the condition with the field along the hard axis[9, 10, 11, 12], the precise field and magnetic axis relationship is difficult to determine since the easy axis lies about 35 degrees away from the c-axis, and the system is, moreover, canted above the spin flop field. Nevertheless, an extrapolation of the AFR resonance to the zero field gives a gap frequency which corresponds (through $h\nu/g\mu_B$), to a characteristic field of 1.7 T. This value, which is related to the product of the exchange field and the anisotropy field, is comparable to the spin-flop field (1.1 T) reported in magnetization studies[5]. We now turn to the non-resonant features in the ac magnetoconductivity measurements, exhibited in Figs. 2, 4, and 5. We have identified three recurrent features in the temperature and field dependent cavity response below T_N which appear regardless of frequency or sample orien-

FIG. 4: Temperature dependence of the cavity response at different fields for $\lambda-(BETS)_2FeCl_4$; $c \parallel \vec{H}$, $a \parallel \hat{r}$, $\nu=$ 64.927 GHz. Three changes in the cavity absorption with temperature are observed at points A, B, and C. Inset: phase boundaries of of A, B, and C with temperature and field.

An important point in our interpretation is that the field and temperature position of the features (A,B,C) are non-resonant, and only weakly dependent on sample orientation. Figure 5 serves to demonstrate these assertions, and also sheds further light on the nature of frequency dependent resonant cavity measurements. Here the sample was measured for three different orientations in field, different frequencies, and different temperatures. The ESR line is prominent and absorptive at two of the frequencies, and for $c \parallel \hat{r}$, $a \parallel \vec{H}$ the AFR signal is observed. Strong ESR or AFR signals are good indications of the sample's coupling to $\vec{H}_{ac}$. (A detailed analysis of cavity mode effects will be published elsewhere[13].) In contrast, the ESR line is weak and inverted for the 71.27 GHz data. Here also the field dependence of the background is reversed. It is apparent that the overall signal depends on how the sample is coupled with an $\vec{E}_{ac}$ or $\vec{H}_{ac}$ excitation mode, as indicated by the intensity of the ESR features. However, the ESR line positions rigourously follow the linear frequency dependence shown in Fig. 3b. Returning to the non-resonant structures, the appearance of these features depends on the cavity geometry and the resonant mode employed. The dissipation of the cavity is governed by the surface resistance, R_s, ($\propto Re\sqrt{\frac{1}{\hat{\sigma}}}$)[14]. The response is a measure of the complex magneto-optical conductivity of the material via the skin depth of the sample, $\hat{\sigma}$. Although different shaped

FIG. 5: Magnetic field dependence of cavity response for $\lambda - (BETS)_2FeCl_4$ in three different sample orientations, frequencies, and temperatures. Arrows indicate the position of the ESR lines, and the AFR line for $a \parallel \vec{H}$ is also indicated. The step-like structures (A,B, C) between 5 and 10 T appear in all cases.

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